Universal Elasticity and Fluctuations of Nematic Gels

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We study elasticity of spontaneously orientationally-ordered amorphous solids, characterized by a vanishing transverse shear modulus, as realized for example by nematic elastomers and gels. We show that local heterogeneities and elastic nonlinearities conspire to lead to anomalous nonlocal universal elasticity controlled by a nontrivial infrared fixed point. Namely, at long scales, such solids are characterized by universal shear and bending moduli that, respectively, vanish and diverge at long scales, are universally incompressible and exhibit a universal negative Poisson ratio and a non-Hookean elasticity down to arbitrarily low strains. Based on expansion about five dimensions, we argue that the nematic order is stable to thermal fluctuation and local heterogeneities down to $d_{lc} < 3$.

Liquid crystal elastomers and gels — weakly crosslinked networks of liquid crystal polymers — combine the electro-optic response and thermodynamic phase behavior of liquid crystals with the mechanical advantages of solids, such as rubber and plastics. They therefore hold considerable technological potential. These materials exhibit rich interplay between orientational order and network elasticity that leads to many unusual properties not found in conventional liquid crystals or in conventional rubber[1]. The most striking of these is the vanishing stress, σ_{ij} in response to a finite strain, u_{ij} applied transversely to the spontaneous[2] uniaxial distortion that develops below the isotropic-nematic (IN) transition. Much progress had been made in understanding these materials both from the neo-classical theory of rubber[1] and from more general symmetry-based elastic formulation[3, 4]. Many of the intriguing properties of nematic elastomers can be traced back to the existence of novel nemato-elastic Goldstone mode[5, 6], associated with a spontaneous breaking of rotational symmetry[3, 4] of the amorphous polymer matrix. Although considerable progress had been made[1], most of the analyses had been limited to mean-field treatments. We have recently developed a fully *nonlinear* elastic theory of nematic elastomers[7], that, for one, allows us to assess the effects of thermal fluctuations[8]. Furthermore, while *statistically* homogeneous and isotropic[2], elastomers are *locally* quite heterogeneous[9]. It is essential to study the role that network heterogeneity plays in determining macroscopic properties of liquid crystal elastomers, and this is the goal of the present Letter.

We find that on scales longer than $\xi^z_{NL} \sim K^2/\Delta$, (with K an effective Frank nematic modulus and Δ a measure of heterogeneity) even arbitrarily weak heterogeneity qualitatively modifies liquid crystal and elastic properties of a nematic elastomer relative to those of the ideal homogeneous and isotropic one[3, 4]. In particular we find that macroscopically a nematic elastomer exhibits a nonlocal elasticity, characterized by shear moduli, that vanish as universal power-laws of system size, implying a host of exotic elastic behavior: (1) a *non*-Hookean elasticity $\sigma_{zz} \sim (u_{zz})^\delta$ (with $\delta > 1$ universal but geometry-dependent), that extends down to arbitrarily weak strains, even along the uniaxial axis $\hat{\mathbf{z}}$[2], contrasting strongly and qualitatively with the *linear* longitudinal stress-strain relation of idealized heterogeneity-free elastomers; (2) a crossover to linear stress-strain relation for strain smaller than a critical strain $u^c_{zz} \sim H^{2/\delta}$ set by an external aligning (e.g., magnetic) H field; (3) a nematic Frank modulus that diverges as a power-law of the minimum of lengths associated with system size, stress or aligning field; (4) universal ratios of elastic moduli, with a negative Poisson ratio in the plane transverse to the nematic axis $\hat{z}$, and (5) a macroscopic incompressibility, independent of microscopic elastic moduli, even for elastomers that are compressible on short scales. In particular, (4) and (5) predict that a strain u_{xx}, applied *transversely* to the nematic axis[10] must be accompanied by strains $u_{yy} = 5u_{xx}/7$ and $u_{zz} = -12u_{xx}/7$, and strain u_{zz}, applied *along* the uniaxial axis will result in $u_{xx} = u_{yy} = -u_{zz}/2$, independent of any other microscopic details.

We also find that because of the compliance of the elastomer network, the nematic order is far more robust to the disordering effects of the local random torques by network's heterogeneities. In fact, a renormalization group analysis and expansion about five dimensions suggests that orientational order of nematic elastomers is stable to weak network heterogeneity[16], in strong contrast to nematics confined in *rigid* gels[11, 12, 13].

Although such quenched disorder and thermally-driven elastic anomalies, controlled by a nontrivial low temperature fixed point have been previously predicted in a variety of other systems[12, 14, 15], to our knowledge, nematic elastomers are a first example of a three-dimensional solid, where these exotic theoretical predictions can be *directly* experimentally tested.

As discussed in great detail in Refs. 3, 4, most of the properties of nematic elastomers can be captured by a purely elastic description in terms of a Lagrangian strain tensor $u_{\alpha\beta} = \partial_\alpha \mathbf{R}\cdot\partial_\beta \mathbf{R} - \delta_{\alpha\beta}$, with the nematic order parameter $Q_{\alpha\beta} = S(\hat{n}_\alpha \hat{n}_\beta - \delta_{\alpha\beta}/3)$ (S and $\hat{\mathbf{n}}$, respectively,

the magnitude and the unit director characterizing the nematic order) integrated out. In this formulation the IN transition is signaled by a spontaneous uniaxial distortion characterized by a strain $u^0_{\alpha\beta} \propto Q_{\alpha\beta}$, that takes place when the effective shear modulus drops below a critical value.

To study the properties of such an orientationally-ordered solid, it is convenient to express its elasticity in terms of the strain tensor (and its gradients) measured relative to the spontaneously uniaxially strained state. The resulting elastic theory resembles that of a conventional uniaxial solid, but with an essential difference, that the shear modulus $\mu_{z\perp}$, associated with distortions transverse to the uniaxial axis identically vanishes[3, 4]. This important feature captures, at the harmonic level, the soft elastic mode associated with the underlying rotational invariance of the high temperature *isotropic* elastomer solid from which the uniaxial state *spontaneously* emerges.

Of course real elastomers are only *statistically* homogeneous and isotropic. As discussed in closely related contexts[12, 14, 15], at long length scales, the elastomer network heterogeneities lead to quenched local random stress $\sigma_{\alpha\beta}(\mathbf{x})$ and anisotropy $\mathbf{g}(\mathbf{x})$ fields[17]

$$\mathcal{H}_R = -u_{\alpha\beta}\sigma_{\alpha\beta}(\mathbf{x}) - \left(\mathbf{g}(\mathbf{x})\cdot\mathbf{n}\right)^2, \tag{1}$$

that locally distort the elastomer relative to an idealized microscopically homogeneous and isotropic state.

As we will show explicitly below, because of the soft mode (vanishing $\mu_{z\perp}$), such random fields (as well as thermal fluctuations) lead to large elastomer distortions at which some of the nonlinear elastic terms become comparable to harmonic ones. It is therefore essential to capture the full underlying (target and reference spaces[4]) rotational invariance (soft mode) of the nematic elastomer. As we have recently demonstrated, this is quite nontrivial and requires a proper treatment of the nonlinear parts of the Lagrangian strain, as well as keeping of the cubic and quartic terms in Lagrange strain tensor[8]. The upshot of that analysis is that for weak heterogeneity, at long length scales elastic and orientational properties of a nematic elastomer[16] are captured by an elastic Hamiltonian

$$\begin{aligned}\mathcal{H} &= \frac{1}{2}\left[B_z w_{zz}^2 + \lambda w_{ii}^2 + 2C w_{zz} w_{ii} + 2\mu w_{ij} w_{ij}\right. \\ &+ \left. K(\nabla_\perp^2 u_z)^2\right] - \sigma(\mathbf{x})\cdot\nabla_\perp u_z,\end{aligned} \tag{2}$$

where

$$\begin{aligned} w_{zz} &= \partial_z u_z + \frac{1}{2}(\nabla_\perp u_z)^2, \\ w_{ij} &= \frac{1}{2}(\partial_i u_j + \partial_j u_i) - \frac{1}{2}\partial_i u_z \partial_j u_z, \end{aligned} \tag{3}$$

are the rotationally-invariant smectic-like and columnar-like nonlinear strain tensors relative to the uniaxial state, i and j are summed over indices x and y transverse to the uniaxial axis z, and $\sigma_i(\mathbf{x}) \approx \frac{1}{2}\sigma_{iz}(\mathbf{x}) + 2g_i(\mathbf{x})g_z(\mathbf{x})$, that we take to be zero-mean Gaussian quenched random field, with spatially independent correlation function

$$\overline{\sigma_i(\mathbf{x})\sigma_j(\mathbf{x}')} = \Delta\delta_{ij}\delta(\mathbf{x}-\mathbf{x}'). \tag{4}$$

Our goal here is to study long scale properties of $\mathcal{H}$. The harmonic correlation functions $\overline{\langle u_\alpha(\mathbf{q})u_\beta(\mathbf{q}')\rangle}_0 = \tilde{G}^0_{\alpha\beta}(\mathbf{q})(2\pi)^d\delta^d(\mathbf{q}+\mathbf{q}')$ of the phonon fields $u_\alpha = (u_i, u_z)$ can be easily calculated,

$$\tilde{G}^0_{\alpha\beta}(\mathbf{q}) = G^0_{\alpha\beta}(\mathbf{q}) + \Delta q_\perp^2 G^0_{\alpha z}(\mathbf{q})G^0_{\beta z}(\mathbf{q}), \tag{5}$$

where $G^0_{\alpha\beta}(\mathbf{q})$ are harmonic correlation functions of an idealized homogeneous and isotropic elastomer, given by

$$G^0_{zz}(\mathbf{q}) = \frac{1}{B_z(1-\rho)q_z^2 + Kq_\perp^4}, \tag{6}$$

$$G^0_{zi}(\mathbf{q}) = -\frac{Cq_z q_i}{(\lambda+2\mu)q_\perp^2(B_z(1-\rho)q_z^2 + Kq_\perp^4)}, \tag{7}$$

$$\begin{aligned} G^0_{ij}(\mathbf{q}) &= \frac{B_z q_z^2 + Kq_\perp^4}{(\lambda+2\mu)(B_z(1-\rho)q_z^2 + Kq_\perp^4)}\frac{q_i q_j}{q_\perp^4} \\ &+ \frac{1}{\mu q_\perp^2}(\delta_{ij} - \frac{q_i q_j}{q_\perp^2}), \end{aligned} \tag{8}$$

and $\rho = C^2/B_z(\lambda+2\mu)$ is a dimensionless ratio. In Eq.5, the second term describes the dominant heterogeneity-induced (frozen) deformations, relative to the ideal nematic elastomer state, with the first giving thermal fluctuations about this ground state.

The validity of the perturbation theory in heterogeneity and elastic nonlinearities is controlled by the fluctuation of the phonon fields. At a harmonic level, at long scales, a representative real-space distortion is given by

$$\overline{\langle u_z(\mathbf{r})^2\rangle} = \text{Const.}\Delta\left(\frac{B_z(1-\rho)}{K^5}\right)^{\frac{1}{2}} L_\perp^{5-d}, \tag{9}$$

The divergence of these distortions with the size of the system $L_{\perp,z}$ for $d < 5$ signals the breakdown of harmonic elasticity on scales longer than nonlinear length scales $\xi_{NL}^{\perp,z}$, which, in 3d are given by

$$\xi_{NL}^{\perp} \approx \frac{K^{5/4}}{(B_z(1-\rho))^{1/4}\Delta^{1/2}}, \qquad \xi_{NL}^{z} \approx K^2/\Delta. \tag{10}$$

To assess the physical consequence of these divergences, we employ a momentum-shell renormalization group (RG), controlled by an expansion in $\epsilon = 5-d$. It is convenient to work with the moduli that renormalize multiplicatively,

$$B = \frac{1}{4}(B_z + B_\perp + 2C), \tag{11}$$

$$\mu_L = \frac{1}{4}(B_z + B_\perp - 2C), \tag{12}$$

$$\tilde{C} = \frac{1}{4}(B_z - B_\perp), \tag{13}$$

where B is the overall bulk modulus, μ_L the longitudinal shear modulus and $\tilde{C}$ the cross coupling. The results of the RG coarse-graining procedure are summarized by flow equations for the effective elastomer parameters on length scale ae^l (a a cutoff set by network's mesh size)

$$\begin{aligned}
\frac{dB}{dl} &= (d+3-3\omega-\eta_B)B,\\
\frac{d\tilde{C}}{dl} &= (d+3-3\omega-\eta_{\tilde{C}})\tilde{C},\\
\frac{d\mu_L}{dl} &= (d+3-3\omega-\eta_L)\mu_L,\\
\frac{d\mu}{dl} &= (d+3-3\omega-\eta_\perp)\mu,\\
\frac{dK}{dl} &= (d-1-\omega+\eta_K)K,\\
\frac{d\Delta}{dl} &= (d+1-\omega+\eta_\Delta)\Delta, \qquad (14)
\end{aligned}$$

where,

$$\begin{aligned}
\eta_B &= \frac{3}{2}\rho_2^2 g_L,\\
\eta_{\tilde{C}} &= \eta_L = \frac{3}{2}g_L,\\
\eta_\perp &= \frac{1}{16}g_\perp,\\
\eta_\Delta &= \frac{8\left(1-\rho_2^2\right)g_L^2+3\left(1+\rho_1+2\rho_2\sqrt{\rho_1}\right)g_L g_\perp}{64\left(1+\rho_1-2\rho_2\sqrt{\rho_1}\right)g_L+96\rho_1 g_\perp},\\
\eta_K &= 2\eta_\Delta+\frac{g_\perp\left(4\left(2+3\,\rho_1-5\,\rho_2\sqrt{\rho_1}\right)g_L+5\,\rho_1\,g_\perp\right)}{4\left(2\left(1+\rho_1-2\,\rho_2\sqrt{\rho_1}\right)g_L+3\,\rho_1\,g_\perp\right)}.
\end{aligned}$$

The physics is controlled by the flows of two dimensionless coupling, $g_L(l)$ and $g_\perp(l)$, and two ratios $\rho_1(l)$ and $\rho_2(l)$ defined as:

$$\begin{aligned}
g_L &= \frac{\Delta\mu_L}{K^3}\sqrt{\frac{K\left(2B-4\tilde{C}+3\mu+2\mu_L\right)}{B(3\mu+8\mu_L)+3\mu(\mu_L+2\tilde{C})-8\tilde{C}^2}}, \qquad (15)\\
g_\perp &= \mu\frac{g_L}{\mu_L}, \qquad (16)\\
\rho_1 &= \frac{\mu_L}{B}, \qquad (17)\\
\rho_2 &= \frac{\tilde{C}}{\sqrt{B\mu_L}}. \qquad (18)
\end{aligned}$$

Flow equations for $\rho_1(l)$ and $\rho_2(l)$ are given by:

$$\begin{aligned}
\frac{d\,\rho_1}{d\,l} &= -\frac{3}{2}g_L\,\rho_1\,(1-\rho_2^2), \qquad (19)\\
\frac{d\,\rho_2}{d\,l} &= -\frac{3}{4}g_L\,\rho_2\,(1-\rho_2^2), \qquad (20)
\end{aligned}$$

where mechanical stability requires $\rho_2 = \tilde{C}/\sqrt{B\,\mu_L} < 1$. Below 5 dimension, we expect elastic nonlinearities to be relevant in the presence of quenched random strains and therefore $g_L(l)$ to flow to a positive finite value. This together with Eqs.19,20 implies that at long scales, ratios $\rho_1(l)$ and $\rho_2(l)$ flow to zero, leading to a considerable simplification of the flow equations for $g_L(l)$ ($\neq 0$) and $g_\perp(l)$

$$\begin{aligned}
\frac{d\,g_L}{d\,l} &= \epsilon g_L-\frac{5g_L\left(64{g_L}^2+176g_L g_\perp+51{g_\perp}^2\right)}{32\left(8g_L+3g_\perp\right)}, \qquad (21)\\
\frac{d\,g_\perp}{d\,l} &= \epsilon g_\perp-\frac{g_\perp\left(-64{g_L}^2+752g_L g_\perp+261{g_\perp}^2\right)}{32\left(8g_L+3g_\perp\right)}. \qquad (22)
\end{aligned}$$

For $d = 3 < 5$, $g_L(l)$ and $g_\perp(l)$ indeed grow at long scales, invalidating harmonic elasticity. Eqs.21,22, with flows displayed in Fig.1, have four fixed points, Gaussian (G), Smectic (S), X, and Elastomer (E), that we list in Table I. It is clear from Eq.2 that for a vanishing

Fixed point	g_L*	$g_\perp*$	η_B	$\eta_L=\eta_{\tilde{C}}$	$\eta_\perp$	η_K	η_Δ
G	0	0	0	0	0	0	0
S	$\frac{4\epsilon}{5}$	0	0	$\frac{6\epsilon}{5}$	0	$\frac{\epsilon}{5}$	$\frac{\epsilon}{10}$
X	0	$\frac{32\epsilon}{87}$	0	0	$\frac{2\epsilon}{87}$	$\frac{35\epsilon}{87}$	$\frac{\epsilon}{58}$
E	$\frac{4\epsilon}{263}$	$\frac{96\epsilon}{263}$	0	$\frac{6\epsilon}{263}$	$\frac{6\epsilon}{263}$	$\frac{106\epsilon}{263}$	$\frac{5\epsilon}{263}$

TABLE I: Fixed point couplings and η exponents for heterogeneous nematic elastomer. $\rho^*_{1,2}=0$ for all fixed points.

shear modulus $\mu = 0$, transverse in-plane phonon modes decouple from u_z. The remaining longitudinal mode is encoded through in-plane density fluctuations w_{ii}, that can be integrated out, leading to a finite shift in B_z, with the model reducing to that of a randomly strained smectic[12]. It is reassuring that this fixed point S, is precisely the same as that found in the study of smectics in aerogel[12]. We are not aware of any physical system that

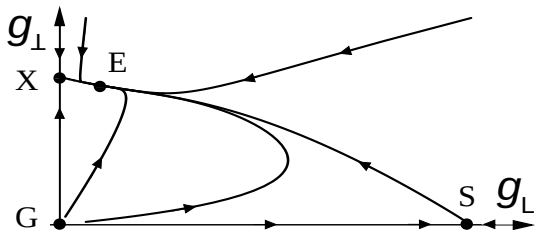

FIG. 1: Flow diagram for dimensionless couplings $g_L(l)$ and $g_\perp(l)$.

is described by the fixed point X, characterized by $g_L = 0$. However, generically elastomers are characterized by a finite in-plane shear modulus μ and a finite modulus μ_L ($g_L \neq 0$), and, as can be seen from Fig.1, S and X are unstable to these couplings. At scales longer than nonlinear crossover lengths $\xi_{NL}^{z,\perp}$, the system flows into a globally stable zero-temperature fixed point E, controlled by elastomer heterogeneities and elastic nonlinearities.

Consequently, at long scales, nematic elastomer can be described by an effectively harmonic but *nonlocal* elastic-

ity, with all elastic moduli, except the overall bulk modulus B length-scale (wavevector) dependent. Standard matching calculation[12] shows that indeed

$$K(\mathbf{k}) = K_0(k_\perp\xi_{NL}^\perp)^{-\eta_K} f_K[(k_\perp\xi_{NL}^\perp)^\zeta/k_z\xi_{NL}^z], \quad (23)$$
$$\mu(\mathbf{k}) = \mu_0(k_\perp\xi_{NL}^\perp)^{\eta_\perp} f_\mu[(k_\perp\xi_{NL}^\perp)^\zeta/k_z\xi_{NL}^z], \quad (24)$$
$$\mu_L(\mathbf{k}) = \mu_0 g_L^*/g_T^*(k_\perp\xi_{NL}^\perp)^{\eta_L} f_{\mu_L}[(k_\perp\xi_{NL}^\perp)^\zeta/k_z\xi_{NL}^z], \quad (25)$$
$$\tilde{C}(\mathbf{k}) = \tilde{C}_0(k_\perp\xi_{NL}^\perp)^{\eta_{\tilde{C}}} f_{\tilde{C}}[(k_\perp\xi_{NL}^\perp)^\zeta/k_z\xi_{NL}^z], \quad (26)$$
$$\Delta(\mathbf{k}) = \Delta_0(k_\perp\xi_{NL}^\perp)^{-\eta_\Delta} f_\Delta[(k_\perp\xi_{NL}^\perp)^\zeta/k_z\xi_{NL}^z], \quad (27)$$
$$B(\mathbf{k}) \approx B_0, \quad (28)$$

where the anisotropy exponent $\zeta = 2 - (\eta_\perp + \eta_K)/2$. Above scaling functions $f_\alpha[v]$ ($\alpha = K, \mu, \mu_L, \Delta$) are v-independent for large v and scale as $v^{\pm\eta_\alpha/\zeta}$ for a vanishing v, such that for $k_\perp \to 0$ limit, the elastic moduli are $k_\perp$-independent. Since this predicts that at long length scale, all elastic moduli are vanishingly small compared to the wavevector-independent bulk modulus B, nematic elastomer is strictly macroscopically *incompressible*, as advertised in the introduction.

Furthermore, from definitions of the coupling g_L and $g_\perp$ we find that the ratio between shear moduli μ and μ_L approaches a universal value $g_\perp^*/g_L^* = 24$ at the fixed point E. This, together with a straightforward analysis and Eq.16 predicts that a uniform strain u_{xx}[10] leads to

$$u_{yy} = \frac{\mu^R - 4\mu_L^R}{\mu^R + 4\mu_L^R}\, u_{xx} \to \frac{5}{7}u_{xx}, \quad (29)$$
$$u_{zz} = -\frac{2\mu^R}{\mu^R + 4\mu_L^R}\, u_{xx} \to -\frac{12}{7}u_{xx}, \quad (30)$$

with negative and positive universal Poisson ratios.

Another fascinating implication of Eqs.23-28 is a strictly nonlinear stress-strain response down to an arbitrarily weak stress $\sigma_{zz} < \sigma_{NL} \equiv K/\xi_{NL,\perp}^2$[2]. To show this we note that σ_{zz} cuts off the singular k dependence (Eqs.23-27) of elastic moduli on scales longer than $\xi_\sigma^\perp = (K\xi_{NL,\perp}^{-\eta_K})^\nu \sigma_{zz}^{-\nu}$, $\nu = 1/(2-\eta_K)$, thereby replacing it by a singular σ_{zz} dependence $\sim (\sigma_{zz}/\sigma_{NL})^{\eta_\alpha\nu}$.[15] In particular we find a non-Hookean response $u_{zz} \sim \sigma_{zz}^{1/\delta}$, $1/\delta = 1 - \eta_L/(2-\eta_K)$, that within (inaccurate) $\epsilon = 2$-expansion gives $\delta = 157/151$ unimpressively close to 1.

Our results rely on the assumption of a stable long-range nematic order[16], requiring convergent orientational fluctuations

$$\overline{\langle|\delta\mathbf{n}|^2\rangle} \approx \overline{\langle|\partial_\perp u_z|^2\rangle} \sim L^{\eta_K+\eta_\perp-2}. \quad (31)$$

Thus a necessary condition for stability of nematic order is $\eta_K + \eta_\perp - 2 < 0$, equivalent to the requirement on the anisotropy exponent $\zeta > 1$. This is satisfied for $d > d_{lc} = 17/56$, suggesting a stability of nematic order for 3d elastomers.

Experimentally, elastomers crosslinked in the isotropic exhibit a polydomain nematic order with a typical micron size correlation length[9, 16]. This can be reconciled with the above prediction of the ϵ-expansion by heterogeneity that is not weak, or by appealing to glassy dynamics that prevents a full equilibration on experimentally relevant time scales. This hypothesis can tested by cooling in the presence of a strong aligning field.

To sum up, we have studied nematic elastomers, taking into account their heterogeneity, and have shown that it leads to many striking elastic properties, that should be readily experimentally testable.

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[16] If in fact the nematic order is unstable (notwithstanding ϵ-expansion prediction), or experiments are done on not fully equilibrated multi-domain samples, we expect our results to continue to hold out to an orientational length ξ_O ($\gg \xi_{NL}$ for $\Delta \to 0$ [12]), beyond which we expect elastomers to exhibit conventional elasticity.
[17] Because elastomer heterogeneity does not break translational symmetry of the target space, unlike liquid crystals confined to aerogel studied in Refs.12, 15, here, no random phonon-field pinning appears.